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PATENT SPECIFICATION

636,423

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PROVISIONAL SPECIFICATION

Improvements in or relating to Adhesive Compositions

We, BERNARD JAMES BALFE, of 12, Needham Road, Stowmarket, in the County of Suffolk, a British Subject, and IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London, S.W.1, a British Company, do hereby declare the nature of this invention to be as follows:—

This invention relates to compositions suitable for bonding purposes, and to methods for adhesively bonding together surfaces by the use of such compositions.

In British Patent Specification No. 581,134 it has been proposed adhesively to unite together materials or articles by the application of a composition comprising an organic diisocyanate modified polyester or polyesteramide, a formaldehyde-liberating substance, an acid or a substance which liberates an acid by interaction with the formaldehyde-liberating substance at room temperatures, and a sufficiency of an organic solvent to render the composition mobile.

According to the present invention, we provide an improved adhesive composition comprising an organic diisocyanate modified polyester or polyesteramide, a terpene-phenolic resin as hereinafter defined, and an organic solvent. We also provide methods for bonding together articles or materials, by the application of such a composition, followed by evaporation of solvent and the bringing together of the surfaces to be united. By the use of the term "terpene-phenolic resin" we mean a condensation product of a terpene with a phenol, or with a phenol and formaldehyde, or with a preformed phenol-formaldehyde condensation product.

The composition may be prepared by simple mixing of solutions and may be used alone or in conjunction with the use of a curing agent for the organic diisocyanate modified polyester or polyesteramide. Another method is to dissolve one component in a solution of the other, or to stir both components together into a solvent or solvent mixture. A further method is to mix the components together, for example in a rubber mill, and then to dissolve the product.

The adhesive compositions employed in this invention are illustrated by the following examples, in which parts are by weight.

EXAMPLE 1

Twenty parts of masticated polyesteramide were dissolved in eighty parts of acetone in a Werner-Pfleiderer mixer. Ten parts of terpene-phenolic resin were added and the whole agitated in a stirrer-mixer until a smooth solution was obtained. The resulting composition was used for attaching nitrocellulose-coated linen fabric and polyvinyl chloride-coated liner fabric to surfaces of wood and metal. Portions of coated fabric, so attached, exposed to ultra-violet light for 24 hours, showed no sign of staining such as will occur with adhesives prepared with phenol-formaldehyde type resins.

EXAMPLE 2

Twenty parts of masticated polyesteramide were dissolved in eighty parts of acetone, and twenty parts of a terpene-phenol resin were then dissolved in the mixture by agitating in a gate-mixer until homogeneity resulted. There was then added 3.3 parts of *pp*'-diphenyl methane diisocyanate and, after mixing, a layer of the resultant adhesive composition was applied to the surfaces of two portions of phenolic mouldings which were to be jointed. The joint was formed by uniting when most of the solvent had evaporated at room temperature (elevated temperature may be employed for accelerating removal of solvent). After 24 hours a strong joint resulted.

EXAMPLE 3

Twenty-five parts of polyesteramide

[Price

were dissolved in a solvent consisting of forty-five parts of acetone, fifteen parts of ethyl acetate and fifteen parts of benzene, solution being assisted by agitation in a stirrer-mixer. Fifteen parts of terpene-phenol resin were then added and the whole mixed until homogeneity resulted. The final adhesive was applied to the outer edge of a phenolic moulded panel and also to an aluminium frame-work. When all the solvent had evaporated the coated articles were clamped together for 24 hours after which a strong joint resulted.

EXAMPLE 4

Polyesteramide was sheeted out on rubber rolls and cut into small pieces following which thirty parts were dissolved in seventy parts of a mixture of acetone, toluene and nitropropane (2:2:1 by weight). Twenty-five parts of a terpene-phenol resin were then mixed in by agitation. To this mixture was added four parts of *pp*'-diphenyl methane diisocyanate. The mixture was then applied to the surface of a low temperature-laminated resin plastic and also to the surface of a steel frame-work. After the most of the solvent had evaporated the plastic and the frame-work were brought together and clamped into position for 24 hours. On removing from the clamps a strong joint was found to have been formed.

The polyesteramide employed in the foregoing Examples was an organic diisocyanate modified polyesteramide, manufactured as described in Example 7 of British Patent Specification No. 580,524, and milled on a rubber mill at 60—70° C. until it was soluble in acetone.

Many methods are available for adhesively uniting surfaces using the compositions of this invention and these are exemplified by the following:—

a) Both surfaces to be united may be coated with the adhesive composition and allowed to get tacky, when they are brought together under pressure.

b) Both coated surfaces may be allowed to dry completely, one or both of them being revived to a tacky state by application of a solvent.

c) The coated surface may be allowed to dry out completely and then united by heat and pressure (using for example electronic heating).

d) As (b) but applying a solution of a curing agent such as *pp*'-diphenyl methane diisocyanate for reviving one or both of the surfaces to a tacky condition.

In addition to the ingredients already mentioned further materials may be incorporated such as, for example, fillers, e.g. carbon black, china clay, asbestos and mica; other resinous materials such as vinyl polymers and copolymers, natural or synthetic rubber and cellulose derivatives.

By the use of this invention strong bonds which are resistant to ageing under hot and humid conditions may be obtained between similar or dissimilar surfaces of woven or other fabrics, for example cotton, cellulose acetate, wool, nylon, paper, regenerated cellulose, wood, metal, leather, polyvinyl chloride sheets, natural and synthetic rubbers and shaped articles or sheets composed entirely or partly of an organic diisocyanate modified polyester or polyesteramide. Thus in the manufacture of footwear the adhesives of this invention may be employed for uniting leather soles or soles composed partly or entirely of polyvinyl chloride, to uppers of leather, woven fabric, coated woven fabric of the kind known as leathercloth, or organic diisocyanate modified polyester or polyester amides, as well as to ornamental shoe parts such as metal brocades. The adhesives may also be employed for bonding abrasive grains to a base material as, for example, in the manufacture of sand-paper and emery-cloth.

Dated the 17th day of September, 1947.

J. W. RIDSDALE,
Solicitor for the Applicants.

COMPLETE SPECIFICATION

Improvements in or relating to Adhesive Compositions

We, BERNARD JAMES BALFE, of 12, Needham Road, Stowmarket, Suffolk, a British Subject, and IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London, S.W.1, a British Company, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

This invention relates to compositions suitable for bonding purposes, and to methods for adhesively bonding together surfaces by the use of such compositions.

In British Patent Specification No.

581,134 it has been proposed adhesively to unite together materials or articles by the application of a composition comprising an organic diisocyanate-modified polyester or polyesteramide, a formaldehyde-liberating substance, an acid or a substance which liberates an acid by inter-action with the formaldehyde-liberating substance at room temperatures, and a sufficiency of an organic solvent to render the composition mobile.

According to the present invention, we provide an improved adhesive composition comprising an organic diisocyanate-modified polyester or polyesteramide, a terpene-phenolic resin as hereinafter defined, and an organic solvent. We also provide methods for bonding together articles or materials by the application of such a composition followed by evaporation of solvent and the bringing together of the surfaces to be united. By the use of the term "terpene-phenolic resin" we mean a condensation product of a terpene with a phenol, or with a phenol and formaldehyde, or with a preformed phenol-formaldehyde condensation product.

The composition may be prepared by simple mixing of solutions and may be used alone or in conjunction with the use of a curing agent for the organic diisocyanate-modified polyester or polyesteramide. Another method is to dissolve one component in a solution of the other, or to stir both components together into a solvent or solvent mixture. A further method is to mix the components together, for example, in a rubber mill, and then to dissolve the product.

The adhesive compositions employed in this invention are illustrated by the following examples, in which parts are by weight.

EXAMPLE I.

Twenty parts of masticated polyesteramide were dissolved in eighty parts of acetone in a Werner-Pfleiderer mixer. Ten parts of terpene-phenolic resin were added and the whole agitated in a stirrer-mixer until a smooth solution was obtained. The resulting composition was used for attaching nitrocellulose-coated linen fabric and polyvinyl chloride-coated linen fabric to surfaces of wood and metal. Portions of coated fabric, so attached, exposed to ultra-violet light for twenty-four hours showed no sign of staining such as will occur with adhesives prepared with phenolformaldehyde-type resins.

EXAMPLE II.

Twenty parts of masticated polyesteramide were dissolved in eighty parts of acetone, and twenty parts of a terpene-phenolic resin were then dissolved in the mixture by agitating in a gate-mixer until homogeneity resulted. There was then added 3.3 parts of *pp'*-diphenyl methane diisocyanate and, after mixing, a layer of the resultant adhesive composition was applied to the surfaces of two portions of phenolic mouldings which were to be jointed. The joint was formed by uniting when most of the solvent had evaporated at room temperature (elevated temperature may be employed for accelerating removal of solvent). After twenty-four hours a strong joint resulted.

EXAMPLE III.

Twenty-five parts of polyesteramide were dissolved in a solvent consisting of forty-five parts of acetone, fifteen parts of ethyl acetate and fifteen parts of benzene, solution being assisted by agitation in a stirrer-mixer. Fifteen parts of terpene-phenolic resin were then added and the whole mixed until homogeneity resulted. The final adhesive was applied to the outer edge of a phenolic moulded panel and also to an aluminium framework. When all the solvent had evaporated the coated articles were clamped together for twenty-four hours after which a strong joint resulted.

EXAMPLE IV.

Polyesteramide was sheeted out on rubber rolls and cut into small pieces following which thirty parts were dissolved in seventy parts of a mixture of acetone, toluene and nitropropane (2:2:1 by weight). Twenty-five parts of a terpene-phenolic resin were then mixed in by agitation. To this mixture was added four parts of *pp'*-diphenyl methane diisocyanate. The mixture was then applied to the surface of a low temperature-laminated resin plastic and also to the surface of a steel framework. After the most of the solvent had evaporated the plastic and the framework were brought together and clamped into position for twenty-four hours. On removing from the clamps a strong joint was found to have been formed.

The polyesteramide employed in the foregoing Examples was an organic diisocyanate-modified polyesteramide, manufactured as described in Example 7 of British Patent Specification No. 580,524, and milled on a rubber mill at 60—70° C. until it was soluble in acetone.

Many methods are available for adhesively uniting surfaces using the compositions of this invention and these are exemplified by the following:—

(a) Both surfaces to be united may be coated with the adhesive composition and allowed to get tacky when they are brought together under pressure.

(b) Both coated surfaces may be

allowed to dry completely, one or both of them being revived to a tacky state by application of a solvent.

(c) The coated surface may be allowed to dry out completely and then united by heat and pressure (using, for example, electronic heating).

(d) As (b) but applying a solution of a curing agent such as *pp'*-diphenyl methane diisocyanate for reviving one or both of the surfaces to a tack condition.

In addition to the ingredients already mentioned further materials may be incorporated such as, for example, fillers, e.g., carbon black, china clay, asbestos and mica; other resinous materials such as vinyl polymers and copolymers, natural or synthetic rubber and cellulose derivatives.

By the use of this invention strong bonds which are resistant to ageing under hot and humid conditions may be obtained between similar or dissimilar surfaces of woven or other fabrics, for example, cotton, cellulose acetate, wool, nylon, paper, regenerated cellulose, wood, metal, leather, polyvinyl chloride sheets, natural and synthetic rubbers and shaped articles or sheets composed entirely or partly or an organic diisocyanate-modified polyester or polyesteramide. Thus in the manufacture of footwear the adhesives of this invention may be employed for uniting leather soles or soles composed partly or entirely of polyvinyl chloride, to uppers of feather, woven fabric, coated woven fabric of the kind known as leathercloth, or organic diisocyanate-modified polyesters or polyesteramides, as well as to ornamnetal shoe parts such as metal brocades. The adhesives may also be employed for bonding abrasive grains to a base material as, for example, in the manufacture of sand-paper and emery-cloth.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. A composition suitable for use as an adhesive comprising an organic diisocyanate-modified polyester or polyesteramide, a terpene-phenolic resin as hereinbefore defined and an organic solvent.

2. A composition substantially as described in any of the Examples.

3. A method of adhesively bonding two surfaces in which a composition according to Claims 1 or 2 is applied to one or both of the surfaces before they are united.

Dated the 7th day of September, 1948.

J. W. RIDSDALE,
Solicitor for the Applicants.

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